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# DEVELOPING TOPOLOGICAL INSULATOR FIBER BASED PHOTON PAIRS SOURCE FOR ULTRAFAST OPTOELECTRONIC APPLICATIONS

NORTHWESTERN UNIVERSITY

*APRIL 2016*

FINAL TECHNICAL REPORT

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**13. SUPPLEMENTARY NOTES**

**14. ABSTRACT**

This report documents research in developing a new source for the production of correlated/entangled photon pairs based on the unique nanolayer properties of topological insulator (TI) materials. The fast nonlinear optical-response of magneto-optic effect together with large third order nonlinearity in TI: $Bi_2Se_3$ offers a singular opportunity for accurate quantum operations in secure quantum communications and quantum computation.

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# Contents

# List of Figures

# 1. Summary

We first studied the nonlinear optical response of a nanolayer topological insulator (TI) by using a short pump pulse to observe the Kerr-nonlinearity effect with an applied magnetic field and to verify the nonlinear response in a pump-probe configuration. We observed pulse broadening and compression due to the magnetic field induced Kerr nonlinearity. These observations are the first step to confirm the existence of Kerr nonlinearity with the applied magnetic field.

# 2. Introduction

This report documents research in developing a new source for the production of correlated/entangled photon pairs based on the unique nanolayer properties of topological insulator (TI) materials. The fast nonlinear optical-response of the magneto-optic effect together with a large third order nonlinearity in TI: $Bi_2Se_3$ offers a singular opportunity for accurate quantum operations in secure quantum communications and quantum computation. The real and imaginary parts of the third-order nonlinearity in TI are responsible for saturable absorption and a nonlinear refractive index, $n_2 = 12\pi^2\chi^{(3)}/n_o c$. The TI: saturation absorber has been developed for Q-switching in lasers at telecom wavelengths [1-3]. The large $n_2 \sim 10^{-10}$ $cm^2/W$ [4] of the TI (compared to ZnO Quantum Dots: $n_2 \sim 10^{-14}$ $cm^2/W$; GaAs: $n_2 \sim 10^{-13}$ $cm^2/W$; Si: $n_2 \sim 10^{-14}$ $cm^2/W$) is relatively unexplored for optoelectronic applications.

The current $\chi^{(2)}$-crystal and $\chi^{(3)}$-fiber based entangled photon source have a limitation on broadband tunability for practical quantum information processing because of the need for pump lasers at specific wavelengths and optical response properties of the nonlinear media.

The topological insulator (TI) [5,6] is a new state of quantum matter with its surface states that has attracted much research in its applications in ultrafast optical devices. The nanolayer of TI provides a gapless surface state consisting of a single Dirac cone. Like graphene, the linear Dirac spectrum dispersion can be engineered for broadband spectral response ranging from terahertz to infrared for optoelectronic applications [7]. In addition, the TI can be made into a broadband tunable Q-switched pump laser, enabling cost-effective fabrication and integration for developing a compact TI photon-pair source with a built-in Q-switched pump laser.

In this report, we first study the nonlinear optical response of a nanolayer topological insulator (TI) by using a short pump pulse with a duration around 220 femtosecond. Our effort is (i) to observe the Kerr-nonlinearity effect with an applied magnetic field and (ii) to verify the nonlinear response by performing a pump-probe configuration. The TI samples are $Bi_2Se_3$ and Fe-doped $Bi_2Se_3$. We observed pulse broadening due to the magnetic field induced Kerr nonlinearity. The pulse broadening in the range from 8 fs – 20 fs is observed with the $Bi_2Se_3$. We observe the pulse compressing of 25 fs with the Fe-doped $Bi_2Se_3$. These observations are the first step to confirm the

existence of Kerr nonlinearity with the applied magnetic field. In addition, we also observe similar effects with other two dimensional crystals such as molybdenum disulfide $MoS_2$. We have also successfully deposited the $Bi_2Se_3$ on an end-facet of an optical fiber.

# 3. Technical Background

In this report, we obtain high quality thin sheets of TI by mechanical exfoliation from $Bi_2Se_3$ bulk crystals which are then dispersed into isopropyl alcohol by ultrasonification. Optical deposition is carried out for placing the nanolayer TI:$Bi_2se_3$ on the end-facet of the single mode fiber patch cord. The deposition process is based on optical trapping force and heat convention effects. The mechanical exfoliation is a very convenient and cost-effective approach which is adopted to synthesize the $Bi_2Te_3$ nanolayer [8].

Even though the large $\chi^{(3)}$-nonlinearity of $10^{-7}$ esu (electrostatic unit) in TI:$Bi_2Se_3$ has been reported, the nanolayer TI: $Bi_2Se_3$ deposited on the end-facet of an optical fiber requires a high peak pump power in order to observe photon pairs through the four-wave mixing (FMW) process. This challenge is overcome by controlling the thickness of TI in optical deposition process.

The surface state of a topological insulator has unusual electronic and optical properties stemming from linear, massless dispersion of electrons near the Dirac point. Magneto-optical properties (Faraday Effect) of thin TI layers have been demonstrated [9]. The large $\chi^{(3)}$-nonlinearity and strong magneto-optical response of TI have attracted a lot of interest for exploring the nonlinear and quantum optical properties of an un-magnetized and magnetized TI and their applications. In this report, we have demonstrated an extremely strong nonlinearity of TI in combination with the magnetic-optical effect.

The thickness of TI and the magnetic-optical effect serve as a systematic approach to tailor $\chi^{(3)}$-nonlinearity leading to nonlinear optical interaction processes such as pulse broadening and compression and the four-wave mixing process. It is also expected that the shifted Landau level [10] at a specific thickness of TI can enhance the $\chi^{(3)}$-nonlinearities through improved phase-matching with Faraday effects and fundamental power concentration, enabling high nonlinearity. We exploit phase matching conditions of nonlinear parametric interaction process in magnetic-optic media [11] for four-wave mixing process in nanolayer TI: $Bi_2Se_3$.

This report provides evidence of magnetic field induced Kerr-nonlinearity and the four-wave mixing process. Since the FMW phase matching condition in TI can be magnetically controlled, a TI platform is potentially capable of generating ultra-fast nonlinear optical control and higher data-rate transmission for information processing in quantum communications than presently available. This is one of the key advantages for the use of TI materials in the generation of non-classical light.

# 4. Methods, Assumptions, and Procedures

## 4.1 Fabrication and Testing of TI: $Bi_2Se_3$

We apply mechanical exfoliation on the bulk TI: $Bi_2Se_3$ by peeling off sheets and flakes as shown in Fig.1. The exfoliated sheet/flake is dispersed into isopropyl alcohol solvent by ultrasonification. Efficient dispersion is achieved after few hours of ultrasonification. The solution is then subjected to centrifugation in order to separate the remaining macroscopic flakes. The visually homogeneous part of the solution is used for the optical deposition process.


**Figure 1 Showing bulk, sheets and flakes of TI: Bi2Se3.**

The experimental setup for an optical deposition is shown in Fig.2. A distributed feedback (DFB) laser source at the wavelength of 1550 nm is spliced to a single mode fiber (SMF) patch cord with a standard FC/PC connector, and then light exiting from the fiber end-facet is directly injected into the dispersion solution. After few minutes of light illumination under a pump power of 40-100 mW, nanolayers of TI:$Bi_2Se_3$ is deposited onto the fiber end-facet. The thickness of the TI:$Bi_2Se_3$ nanolayer is controlled by the average power of the DFB laser and deposition times.

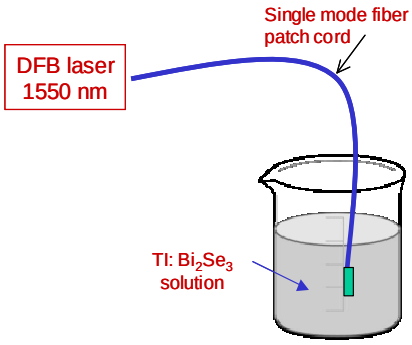


**Figure 2 Optical deposition experiment.**

## 4.2 Stimulated Four-Wave Mixing Process (A Pump-Probe Experiment)

### 4.2.1 Pulsed pump and pulsed signal probe experiment with magnetic field

We prepared a few 10 nm thickness of TI:$Bi_2Se_3$ nanolayer on the end-facet of the SMF for verifying $\chi^{(3)}$-nonlinearity through a pump-probe configuration. Since the value of $\chi^{(3)}$-nonlinearity of TI: $Bi_2Se_3$ is around $10^{-7}$ esu (electrostatic unit) or the $n_2 \sim 10^{-10}$ $cm^2/W$ or nonlinear coupling coefficient $\gamma \sim 10^5$ $W^{-1}km^{-1}$, we use high peak pump power and idle-probe signal for the stimulated four-wave mixing process in the few nanolayers TI:$Bi_2Se_3$. We obtain the pulsed pump and signal light from an IMRA femtosecond mode-locking

fiber laser by using a wavelength-division-multiplexer (WDM) and a tunable Newport 1-nm filter as shown in Fig.3.

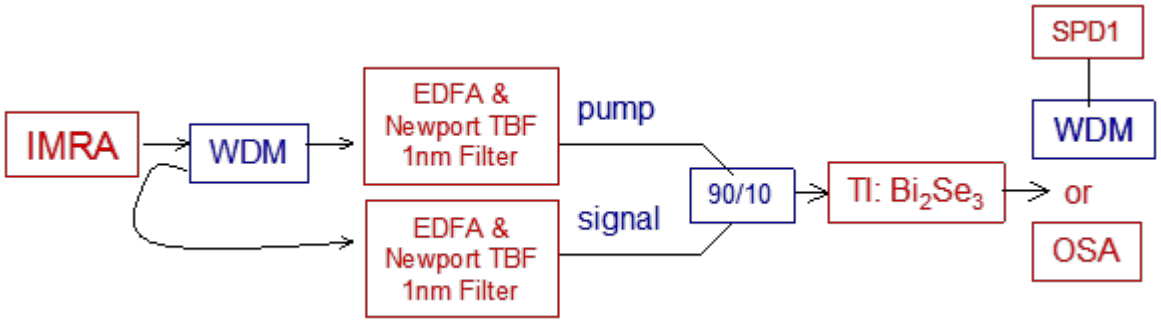


**Figure 3 Setup for the stimulated four-wave mixing process with the pulsed pump and pulsed signal probes. IMRA: femtosecond mode-locking fiber laser, WDM: Wavelength-division multiplexing, EDFA: Erbium-Doped Fiber Amplifier, SPD: single photon detector, OSA: opt**

Two Erbium-Doped Fiber Amplifiers (EDFA) are used for increasing the peak powers of the pump and the idler-probe so that the generated signal is observed in an optical spectrum analyzer (OSA) and photodiode detector.

When the magnetic field is applied normal to the nanolayers TI: $Bi_2Se_3$ and parallel to the propagation direction of the pump, the giant $\chi^{(3)}$-nonlinearity is obtained through the shifted Landau level [10] and the phase matching condition is enhanced by using Faraday Effect.

In a FWM process, two pump photons at frequency of $\omega_3$ are converted to signal and idler photons at respective frequencies $\omega_1$ and $\omega_2$ such that $2\omega_3 = \omega_2 + \omega_1$. The gain of the signal and idler photons depends on the phase mismatch due to the linear dispersion of their propagation constants $\Delta k = 2k_3 - k_2 - k_1$ and the Faraday effect on their angular momenta $\Delta\alpha = 2\alpha_3 + \alpha_2 + \alpha_1$. Here $\alpha_i$ (i=1,2,3) is given by $\omega_i \Theta_i/(2\, n_i\, c)$, where $\Theta_i$ is the specific Faraday rotation due the applied magnetic field in the TI: $Bi_2Se_3$. To obtain a high production rate of photon pairs (signal/idler photons), we must satisfy the condition $\Delta k = \Delta\alpha$. We have adopted the phase matching condition of the parametric interaction process in magneto-optic media [11] similar to the nanolayer TI: $Bi_2Se_3$. This phase matching condition is fulfilled in nanolayer TI: $Bi_2Se_3$ with the pump in right-circularly polarized light RCP (LCP) and the signal and idler both in the left-circularly polarized light LCP (RCP), respectively.

# 5. Results and Discussion

Below is the final result of the project. We obtained topological samples ($Bi_2Se_3$ and Fe-doped $Bi_2Se_3$) from our collaborator Prof. Y. S. Hor from Missouri University of Science and Technology. Our first demonstration is to measure the effect of magnetic field induced Kerr nonlinearity on the temporal profile of a pulse width on a femtosecond time scale. We apply a magnetic field of about 3 tesla normal to the surface of the sample. We observe the pulse broadening (8-20 fs) and compressing (25 fs) in the nanolayer topological insulator. The temporal profile of a pump pulse is measured by using a Mach-Zehnder interferometer. We also confirm the pulse broadening effect by using a two dimensional crystal layer such as molybdenum disulfide $MoS_2$. After we confirmed the existence of the magnetic field induced Kerr-nonlinearity on an optical pulse in the time domain, we proceeded to verify the four-wave mixing process in the topological insulator and molybdenum disulfide by using a pump-probe configuration. In addition, were able to observe the four-wave mixing process in $MoS_2$ in the forward degenerate four-wave mixing at the pump wavelength of 780 nm.

## 5.1 The Overall Project Accomplishment

### 5.1.1 Sample: Topological Insulator

A) $Bi_2Se_3$

B) Fe doped $Bi_2Se_3$

**Figure 4 Samples of $Bi_2Se_3$ and Fe-doped $Bi_2Se_3$ with different thickness, respectively**

After we cleave a thin layer from a bulk crystal of topological insulator (TI), we place the thin TI on a white paper with a square hole. We prepare few different layers with different thicknesses and attach on the ultra-thin glass slide. The glass slide is then mounted on a mirror mount and placed normal to the applied magnetic field as shown in Fig.5. Fig. 4, A and B show the samples of $Bi_2Se_3$ and Fe-doped $Bi_2Se_3$ with different thickness, respectively.

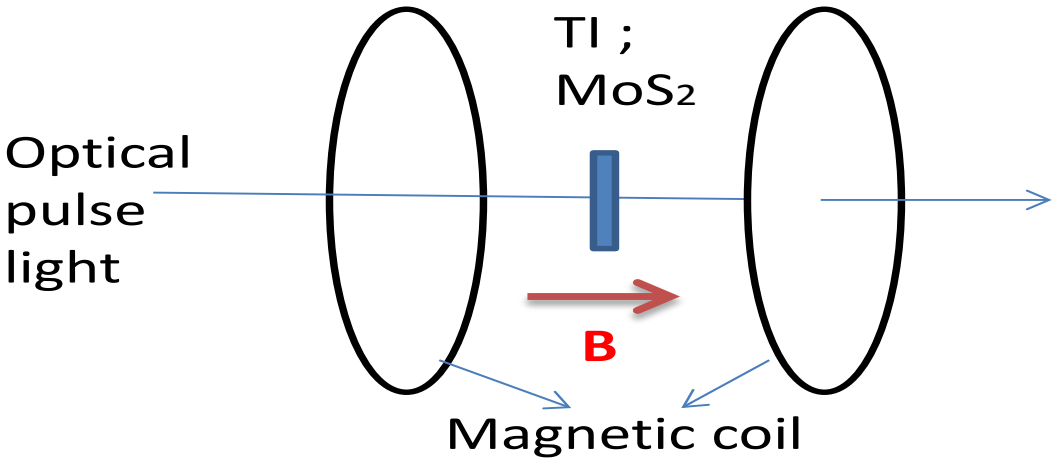


**Figure 5 The sample is placed normal to the applied B field and optical beam.**

### 5.1.2 Sample: Molybdenum Disulfide

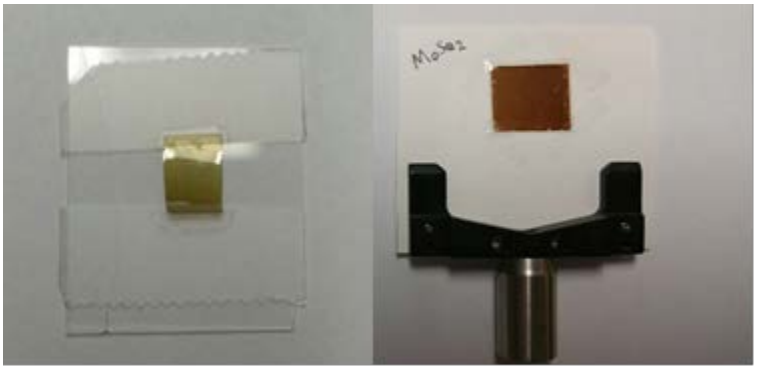

**Figure 6 The MoS2 (left) is deposited on a polymer film PMMA. The MoS2 (right) is deposited on an ultra-thin glass slide.**

### 5.1.3 Pulse Broadening and Compression

We setup an interferometer as shown in Fig.7. The sample with magnetic coil is placed in one arm of the interferometer. The laser pulse light at 780 nm with a pulse duration around 220 fs is used to measure the nonlinear pulse broadening and compression due the magnetic field induced Kerr-nonlinearity.

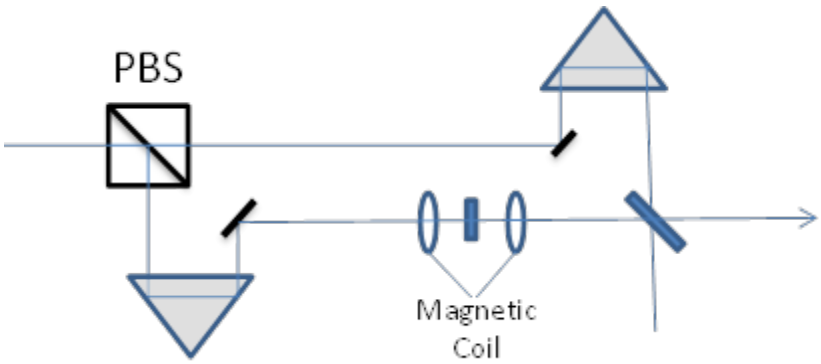


**Figure 7 The interferometer setup for measuring the pulse duration.**

1). **$Bi_2Se_3$**. We measure the pulse duration of a thin layer of topological insulator $Bi_2Se_3$ with the transmission of T = 50%. We apply magnetic field B=3 tesla normal to the sample and parallel to the propagation direction of an optical beam. Without the applied B field, the pulse width after the light pulse passing through the sample is about 260 fs as shown in Fig. 8(a). With the applied B field, the pulse width becomes broaden to 267 fs as shown in Fig. 8(b).

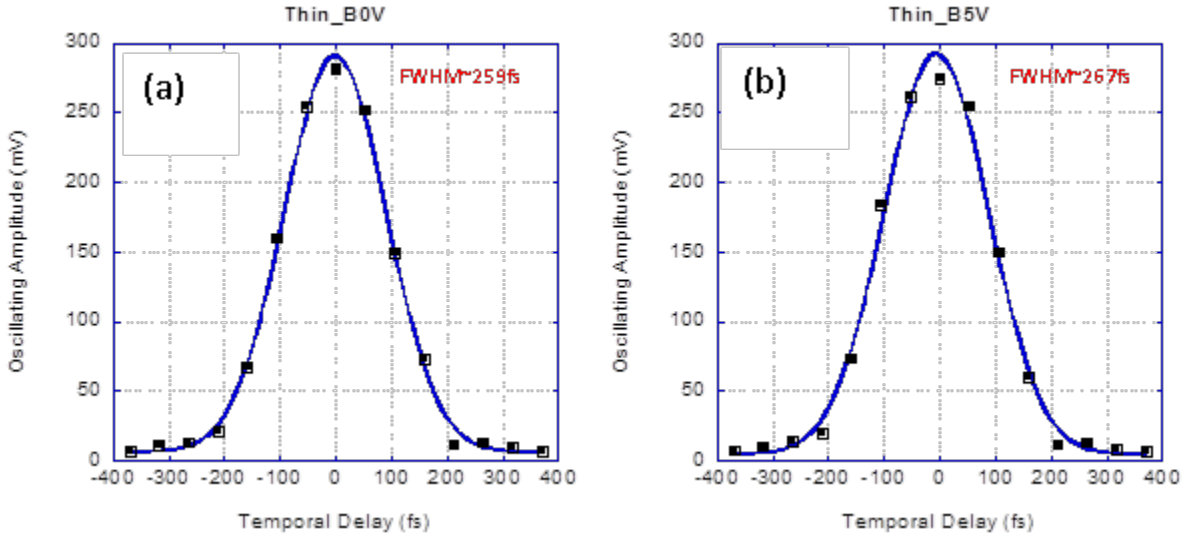


**Figure 8 (a) The FWHM of the pulse duration of 260 fs without B field. (b) The FWHM of the pulse duration of 267 fs with B field.**

We perform the similar measurement on the thick sample with the transmission of 5%. We use a lens to collect the light pulse after the interferometer. The pulse duration with the lens without a sample is about 248 fs as shown in Fig.9. We then insert the sample and measure the pulse duration with and without the applied magnetic field B. We obtain the pulse broadening from 244 fs (B=0 tesla) to 265 fs (B=3 tesla) as shown in Fig.10 and 11.

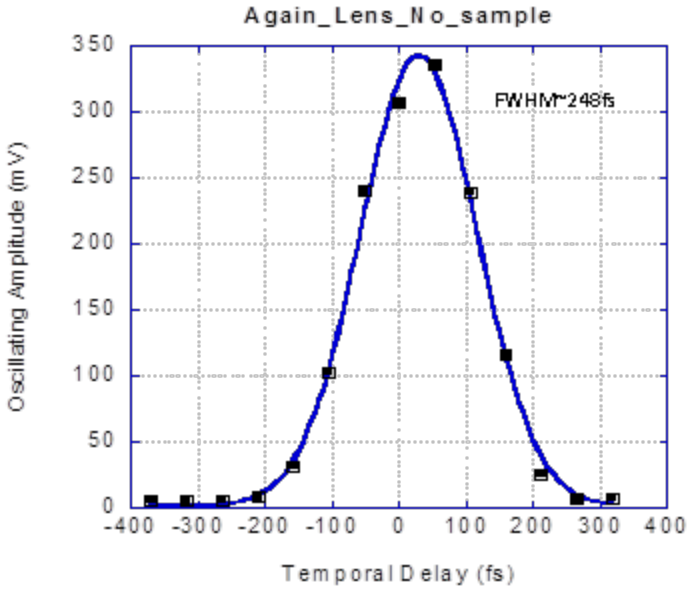


**Figure 9 FWHM of the pulse duration with a lens but no sample.**

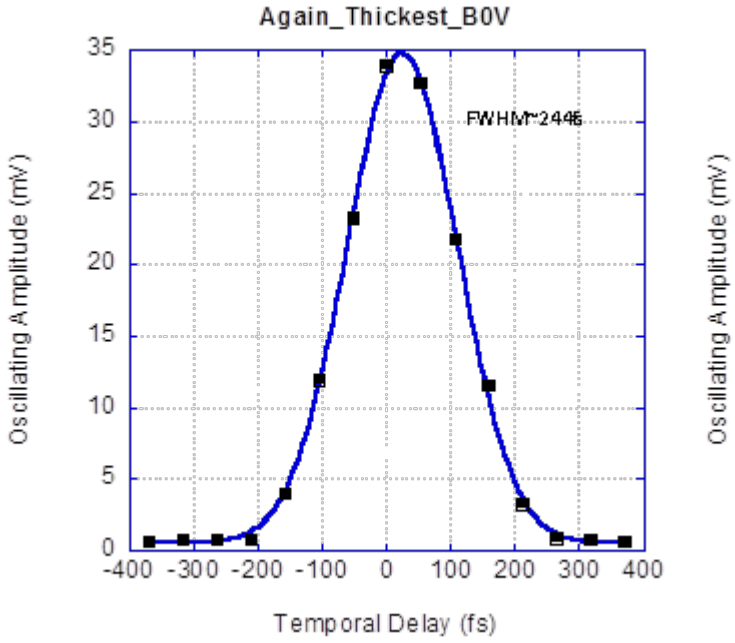


**Figure 10 FWHM of the pulse duration of 244 fs with a lens and a sample but no B field.**

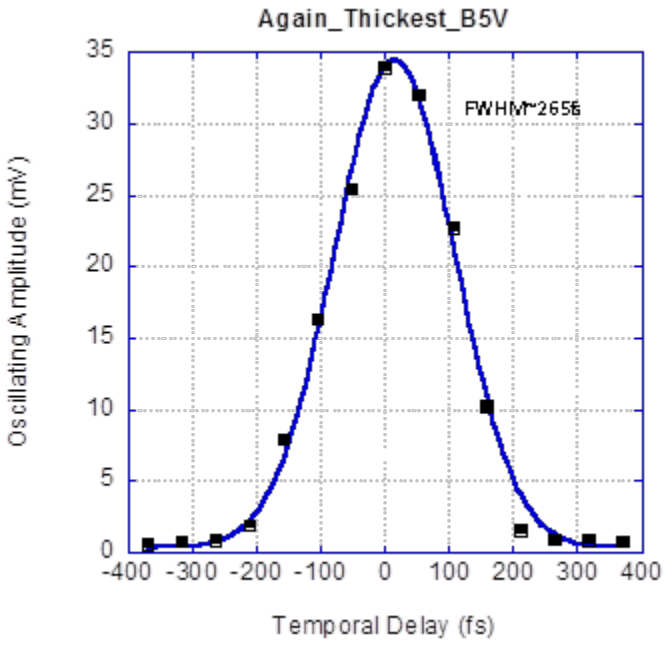


**Figure 11 FWHM of the pulse duration of 265 fs with a lens and a sample with B=3 tesla.**

2) **Fe-doped $Bi_2Se_3$**. We perform the similar measurement on another sample Fe-doped $Bi_2Se_3$ with a transmission of 10%. We use a lens to collect the light pulse after the interferometer. The pulse duration with the lens without the sample is about 276 fs as shown in Fig.12. Then, we observe the pulse compression from 276 fs to 249 fs with the Fe-doped $Bi_2Se_3$ as shown in Fig.13(a) and (b). The applied magnetic field did not affect the pulse compression on the sample.

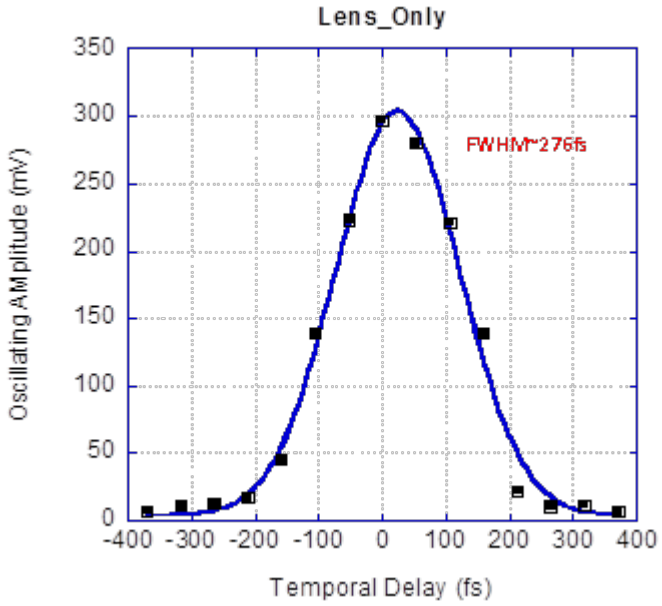


**Figure 12 FWHM of the pulse duration with a lens but no sample.**

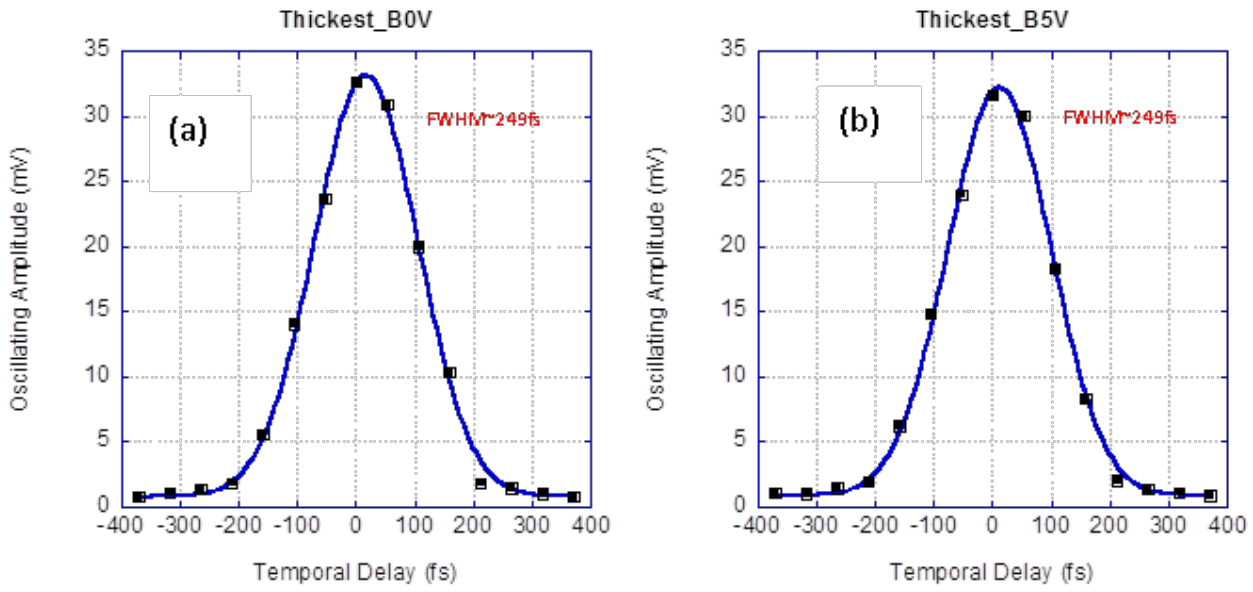


**Figure 13 (a) The FWHM of the pulse duration of 249 fs without B field. (b) The FWHM of the pulse duration of 249 fs with the B = 3 tesla.**

3) **Molybdenum Disulfide ($MoS_2$)**. We also perform a similar measurement on $MoS_2$, which is a two-dimensional crystal. The sample had a transmission of about 80%. We did not use a lens for collecting the light pulse after the interferometer. The FWHM of pulse duration with and without B field are 200 fs and 220 fs, respectively, as shown in Fig. 14(a) and (b).

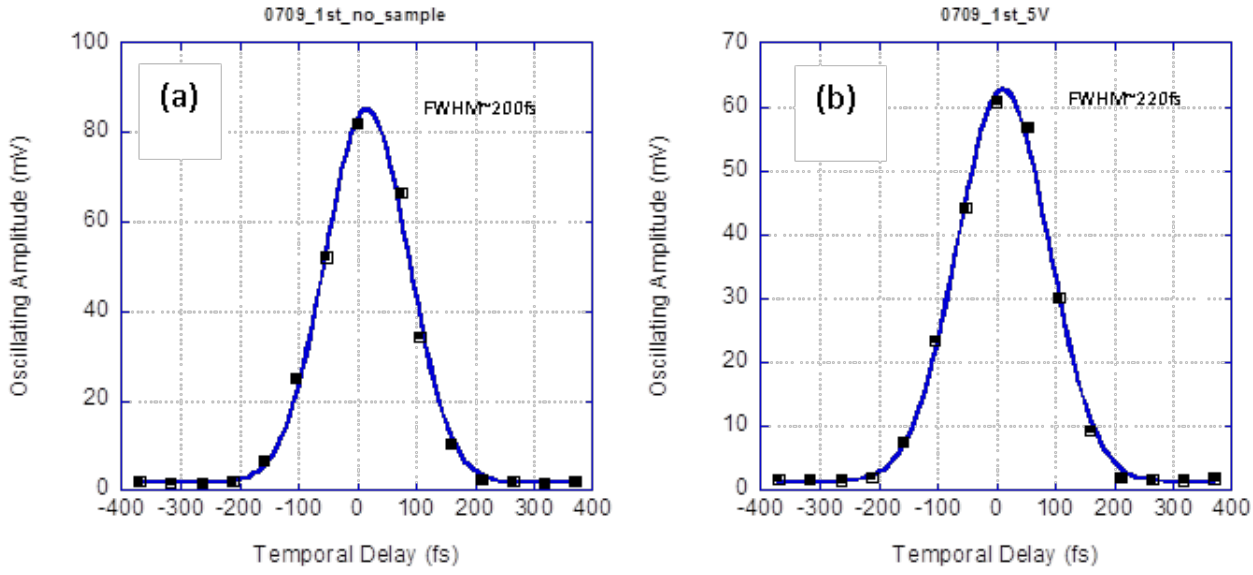


**Figure 14 (a). Without the MoS2, the FWHM of the pulse duration is 200 fs. (b) The FWHM of the pulse duration is 220 fs with the sample MoS2 and B = 3 tesla.**

### 5.1.4 Beam Diffraction

We also observe beam diffraction when an optical light beam is passing through $MoS_2$ sample in Polymethylmethacrylate (Acrylic) PMMA. We did not apply a magnetic field for the beam diffraction measurement. We use a screen, Fig. 15, placed 2 meters away to measure the beam diffraction after the sample.


**Figure 15 Screen**

Fig. 16A shows the location of the optical beam on the screen without the sample. With the sample, the beam is shifted to an angle of about 0.2º as shown in Fig. 16B. When we displace the sample from the right to the left, the light beam is deflected with an angle as large as 0.4º, Fig. 17 A,B.

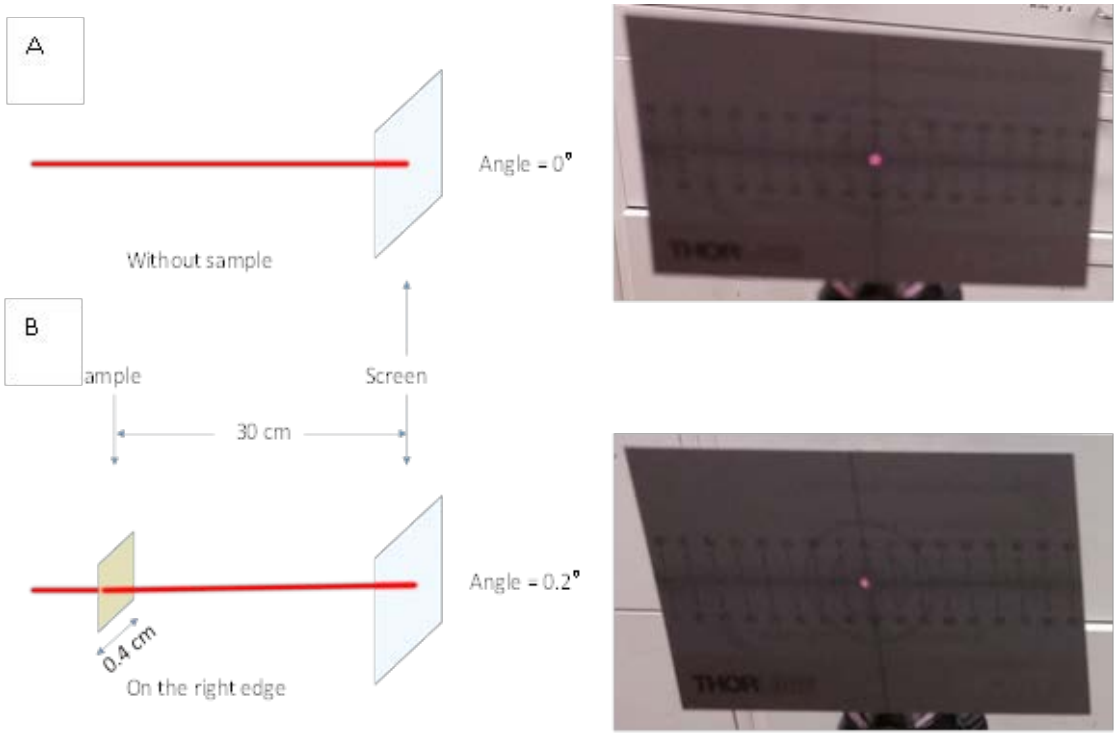


**Figure 16 Optical beam location with/without sample**

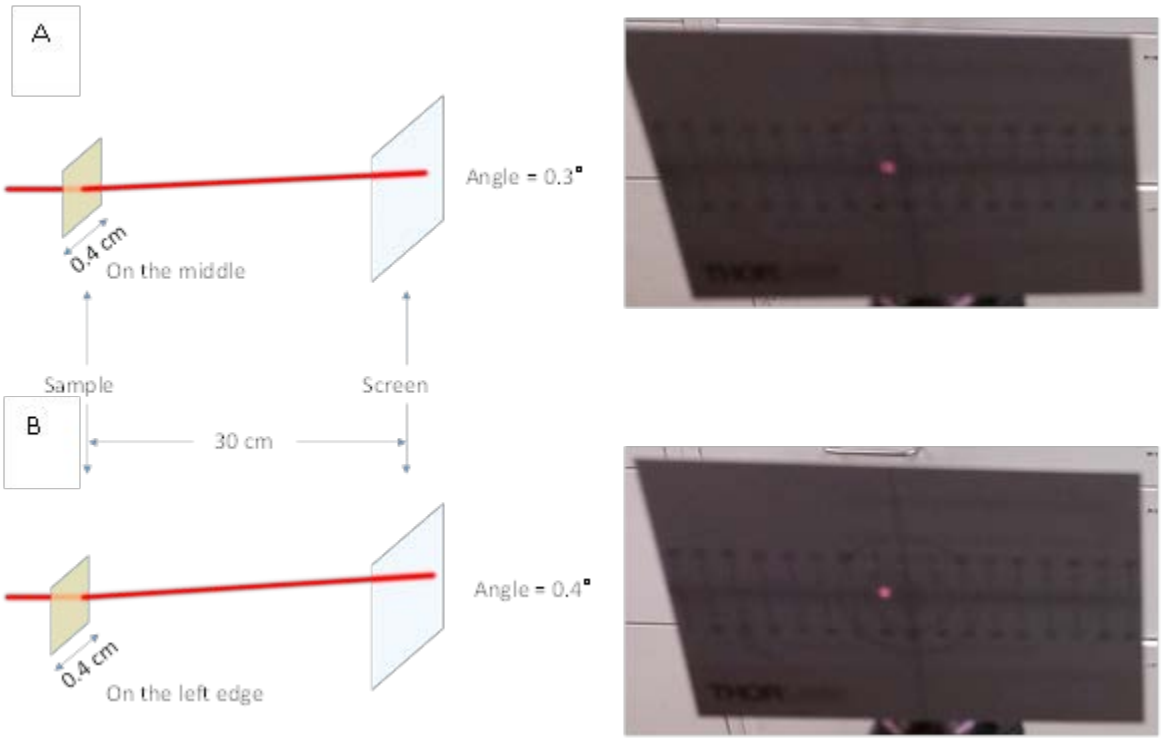


**Figure 17 Optical beam location with displacing the sample from the right to the left.**

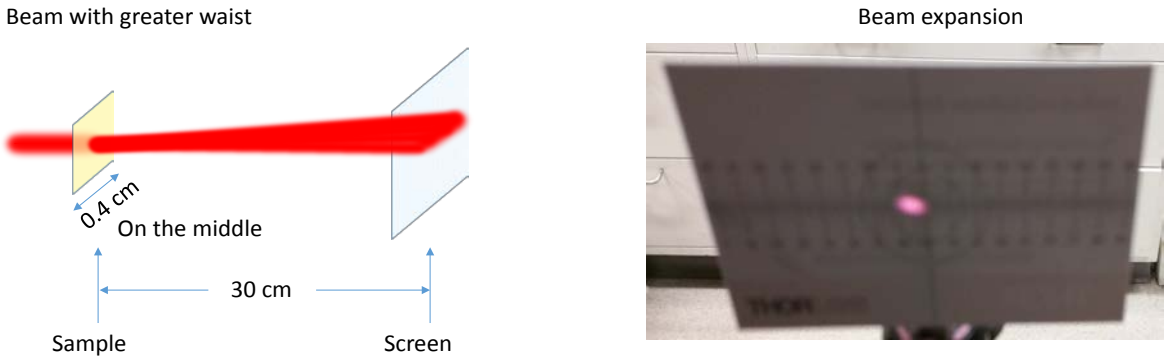


**Figure 18 Large optical beam size**

We then used a large optical beam size for studying the beam diffraction. Fig.18 shows that the large optical beam is deflected and spread to the left.

We believe that the observation of the beam diffraction is caused by the cascaded $\chi^{(3)}$ - and $\chi^{(2)}$ – nonlinearities as observed in Reference [12].

## 5.2 Pump-Probe Experiment

After we confirmed that there is a Kerr-nonlinear effect in the topological insulator $Bi_2Se_3$, we setup a pump-probe configuration for observing the four-wave mixing process.

The experimental setup is shown in Fig.19. The pump is at the wavelength of 1554 nm and the idler probe is at the wavelength of 1559 nm.

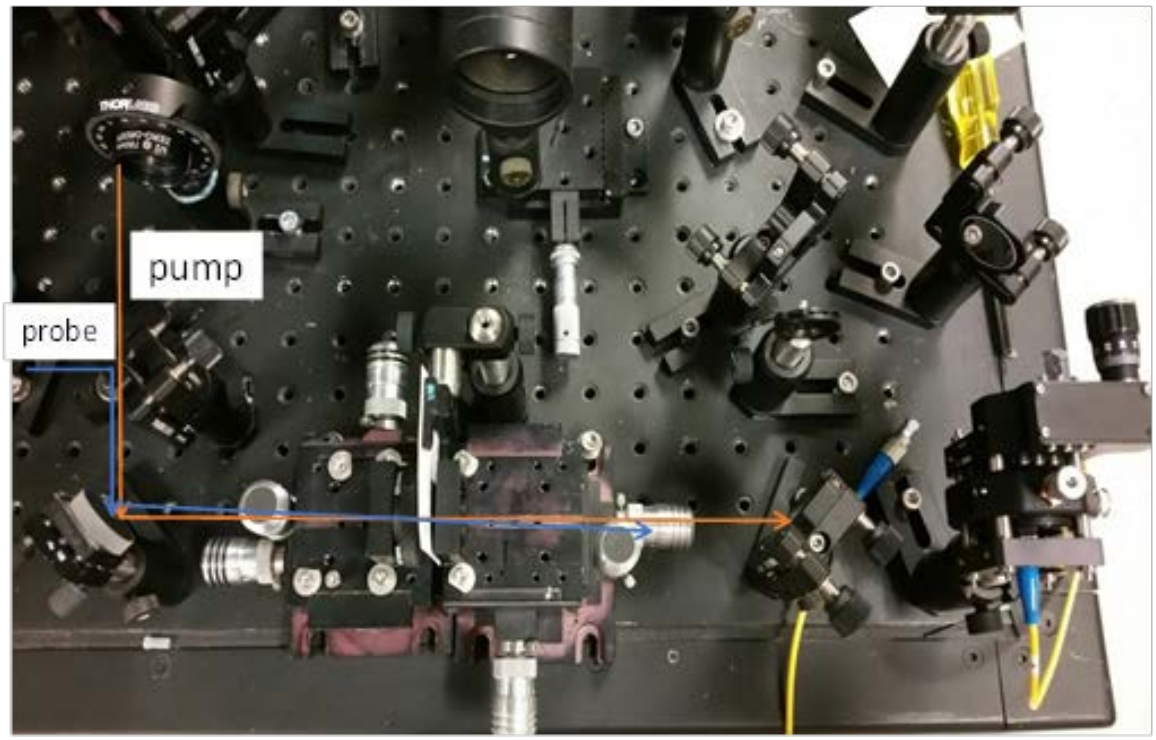


**Figure 19 The pump-probe setup for verifying the four-wave mixing process in topological insulator Bi2Se3 and also the MoS2.**

The generated signal is at the wavelength of 1549 nm. We measured the generated signal by using a photodiode detector as shown in Fig.20. We first measure the pump scattered into the photodiode detector as shown in Fig. 20(a). It shows the voltage of 85 mV. Then, we measure the generated signal and the scattered pump as shown in Fig. 20(b). It shows a voltage of 100 mV. Fig. 20(c) shows the voltage of the scattered idler probe of 4 mV. Using the simple algebra, we obtain the generated signal as 100 mV-85 mV- 4 mV= 11 mV. Our finding has confirmed that the four-wave mixing process occurs in topological insulator $Bi_2Se_3$ and Molybdenum Disulfide $MoS_2$.

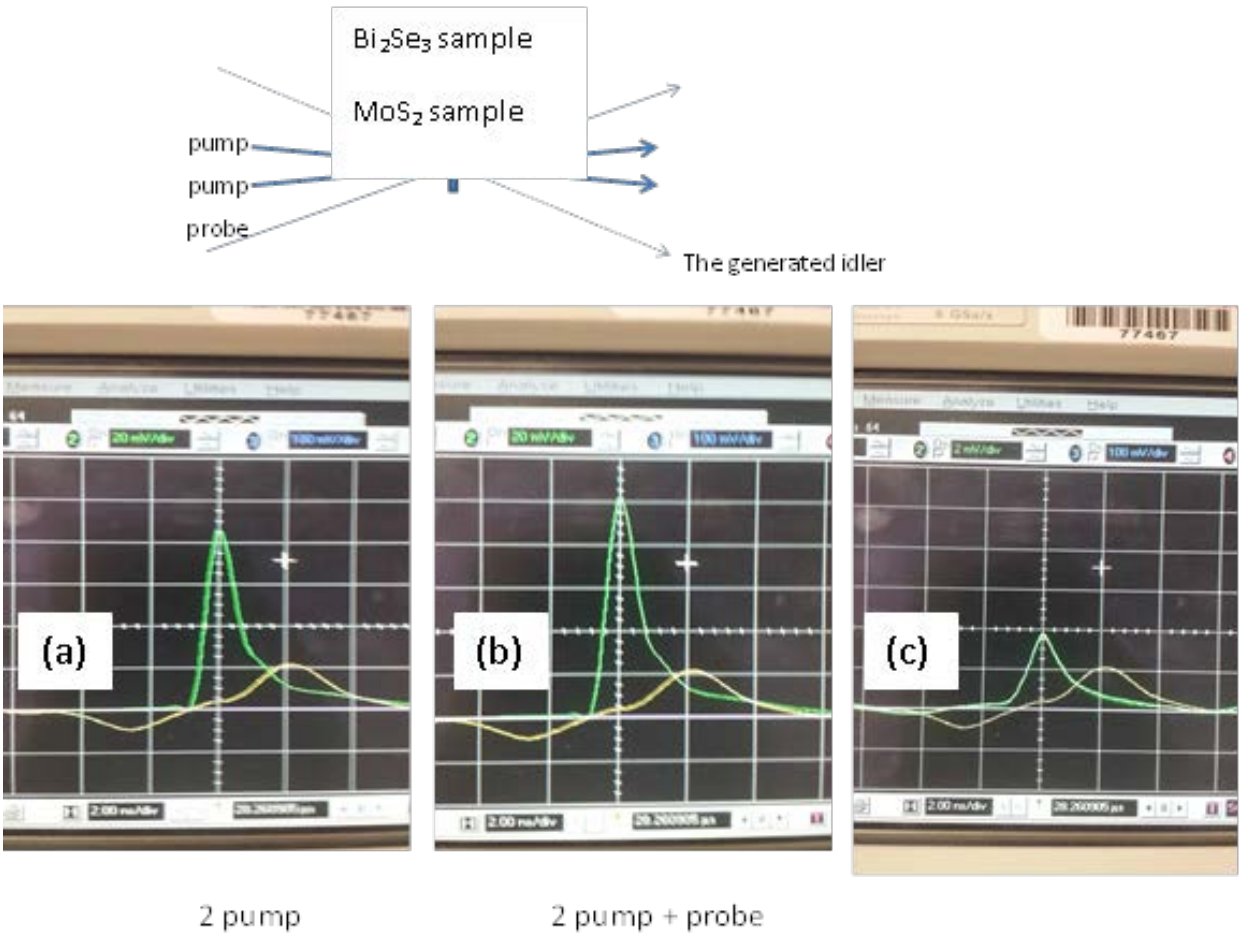


**Figure 20 The voltage of photodiode detector for (a) the scattered pump light, (b) the scattered pump light and the idler-probe light and the generated signal light, and (c) the scattered idler-probe light.**

# 6. Conclusions

We have observed the effect of Kerr-nonlinear induced by magnetic field in the Topological Insulator $Bi_2Se_3$ and Molybdenum Disulfide $MoS_2$. The nonlinear effect is pulse broadening, compression and beam diffraction. We also confirm the four-wave mixing process in topological insulator and $MoS_2$ by using a pump-probe configuration.

# 7. References

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